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IRRADIATION STUDIES ON NERVA FUEL MATERIAL

CONTRACT YEAR 1963

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I. Introduction

Studies of the irradiation behavior of NERVA fuel material are being conducted in steady-state and transient irradiations in the General Electric Testing Reactor (GETR) and in the Transient Test Reactor (TREAT), respectively. The evaluation of NERVA fuel at equivalent full power operation (2-4 kw/cc) requires thermal neutron fluxes of 3-6 x 10^{14} n/cm^2 x sec for times up to 30 minutes. This combination of flux and time is not available in existing irradiation facilities. The reactors used for the program bracket the two requirements. The GETR provides the required time and 20% of the neutron flux requirement, TREAT provides the neutron flux for periods up to 15 seconds. GETR can be used to obtain preliminary steady-state, high-temperature fuel material behavior with respect to fission product release, dimensional stability and fuel migration. On the other hand, TREAT is useful to study the effects of thermal stress on fueled graphite matrix behavior.

II. Transient Irradiation Experiments (TREAT)

A. Experimental Techniques

Transient irradiations were carried out in Argonne National Laboratory's TREAT reactor at the National Reactor Testing station in Idaho. TREAT is a homogeneous, graphite-moderated and graphite-reflected, transient nuclear reactor. The core is composed of graphite-urania matrix fuel elements encased in zircaloy cans. The overall dimensions of each fuel element including reflector graphite are 4 in. x 4 in. x 8 ft. The reactor fuel has a carbon to uranium-235 ratio of 10,000:1. The large amount of carbon serves as moderator, as a large heat sink and provides a negative temperature coefficient of reactivity. The reactor is capable of developing transient thermal flux integrals of approximately 3.5 x 10^{15} nvt thermal. The maximum energy release of the reactor is 1000 Mw-sec with a period of 40 milliseconds. (See Figure 1)

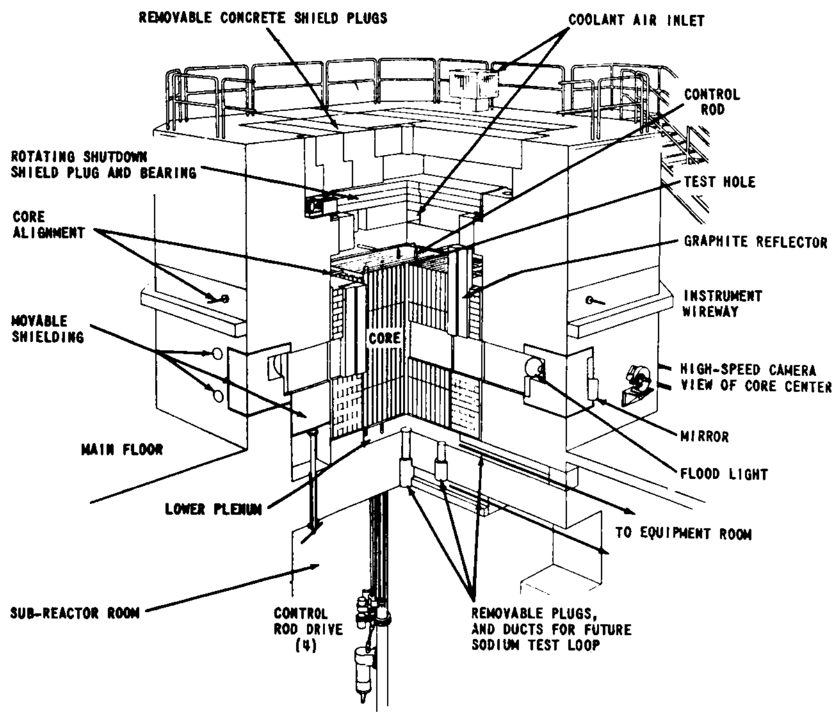

FIGURE 1 - CUTAWAY ISOMETRIC OF TREAT REACTOR

VIEW OF COMPONENTS

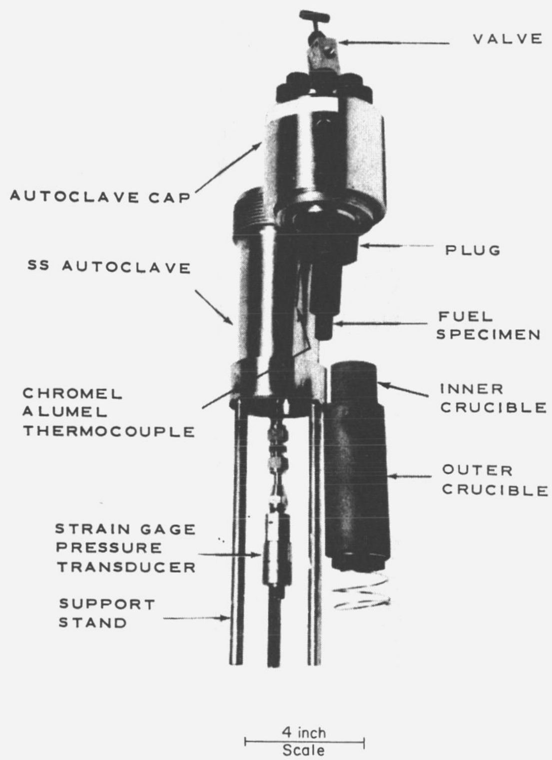

FIGURE 2 - AUTOCLAVE ASSEMBLIES FOR TESTING GRAPHITE FUEL IN TREAT

SCHEMATIC OF THE MODIFIED TRANSPARENT CAPSULE

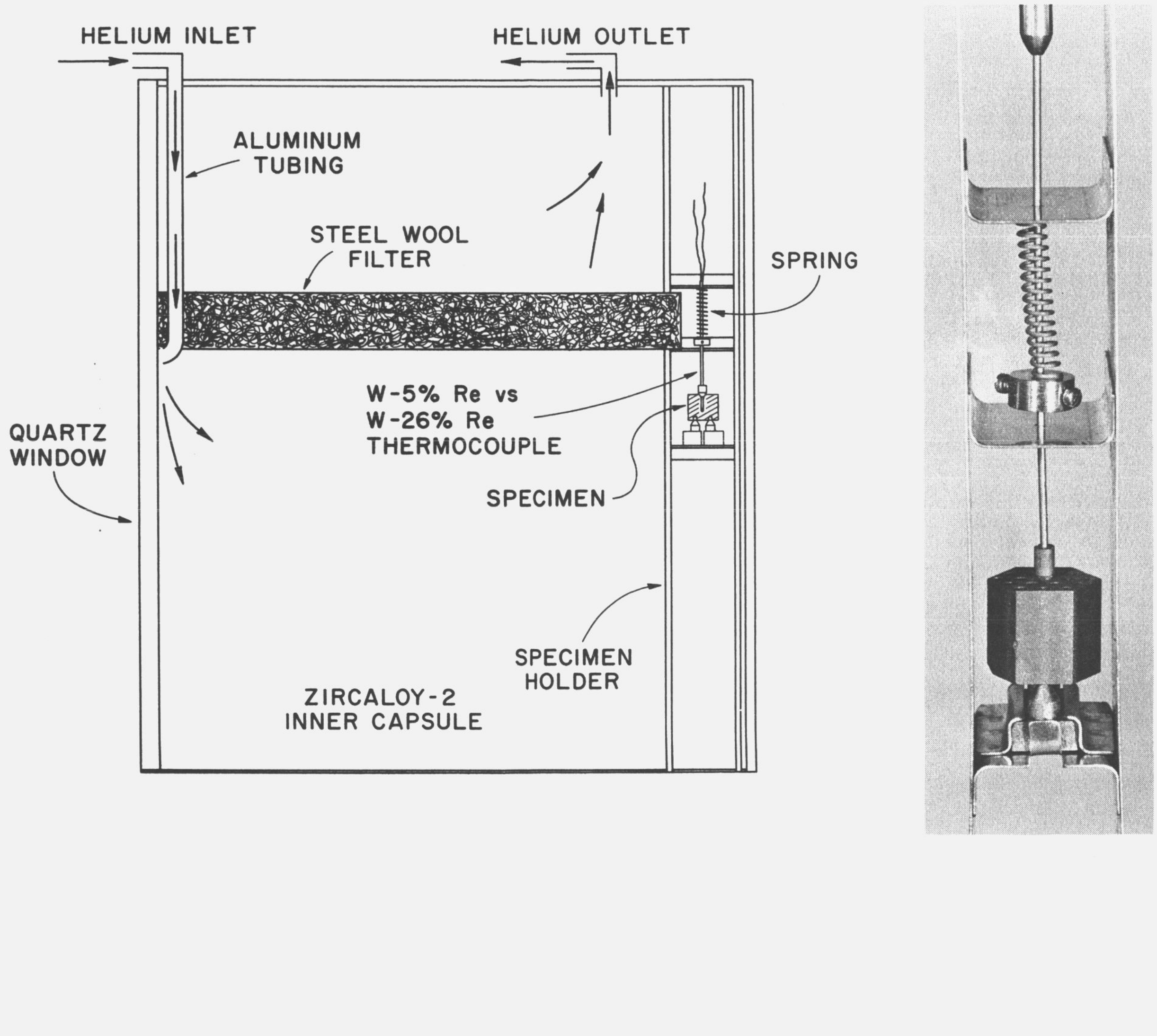

FIGURE 3 - CAPSULE AND FUEL ARRANGEMENT FOR PHOTOGRAPHIC EXPERIMENTS

Two types of experimental assemblies were used to carry out the transient irradiations. The first type was a high pressure stainless steel autoclave which was placed in a dummy TREAT element in the center of the reactor (See Figure 2). A second type of assembly was used for motion picture studies in TREAT. This assembly consisted of a low pressure zircalloy capsule containing a transparent window. The fuel specimen was viewed through a series of mirrors. This arrangement provided means for high speed motion picture photography and for measuring fuel surface temperature with a high response optical pyrometer. (See Figure 3)

The experimental procedure used for each transient pulse consisted of bringing the reactor to a nominal steady-state power level of 10 w. Excess reactivity is then added above prompt critical to initiate the pulse. The total energy of the pulse is temperature limited by the negative temperature coefficient of the TREAT fueled graphite elements, or the pulse can be clipped by rapidly inserting control rods. The period and peak power of a pulse can be controlled by the addition of the proper excess reactivity. Thus, it is possible to exercise control over both the rate of energy input (period) and the total energy input (integrated power). Fuel sample heat-up during fast pulses (less than 75 ms period) is essentially an adiabatic process. Any desired fuel sample temperature (temperatures greater than 3500°C have been attained) may be achieved by selecting the proper value of integrated power for a given size fuel sample and for a particular capsule arrangement.

B. Calculations of Temperature Distribution

Manual calculations were carried out to establish fuel temperature profiles during cooling cycles from elevated temperatures. The model used assumed radiation heat loss from the surface of an infinitely long cylinder having the same cross-sectional area as the fuel hexagon. The calculations assumed adiabatic fuel heating during the transient heat-up, followed by radiation cooling after the peak temperature was attained. The calculations employed the method developed for the quenching of metal shapes*. Figure 4 is a plot of maximum temperature difference between center and surface developed during

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* J. B. Austin, "The Flow of Heat in Metals", Chapter IV, ASM 1942

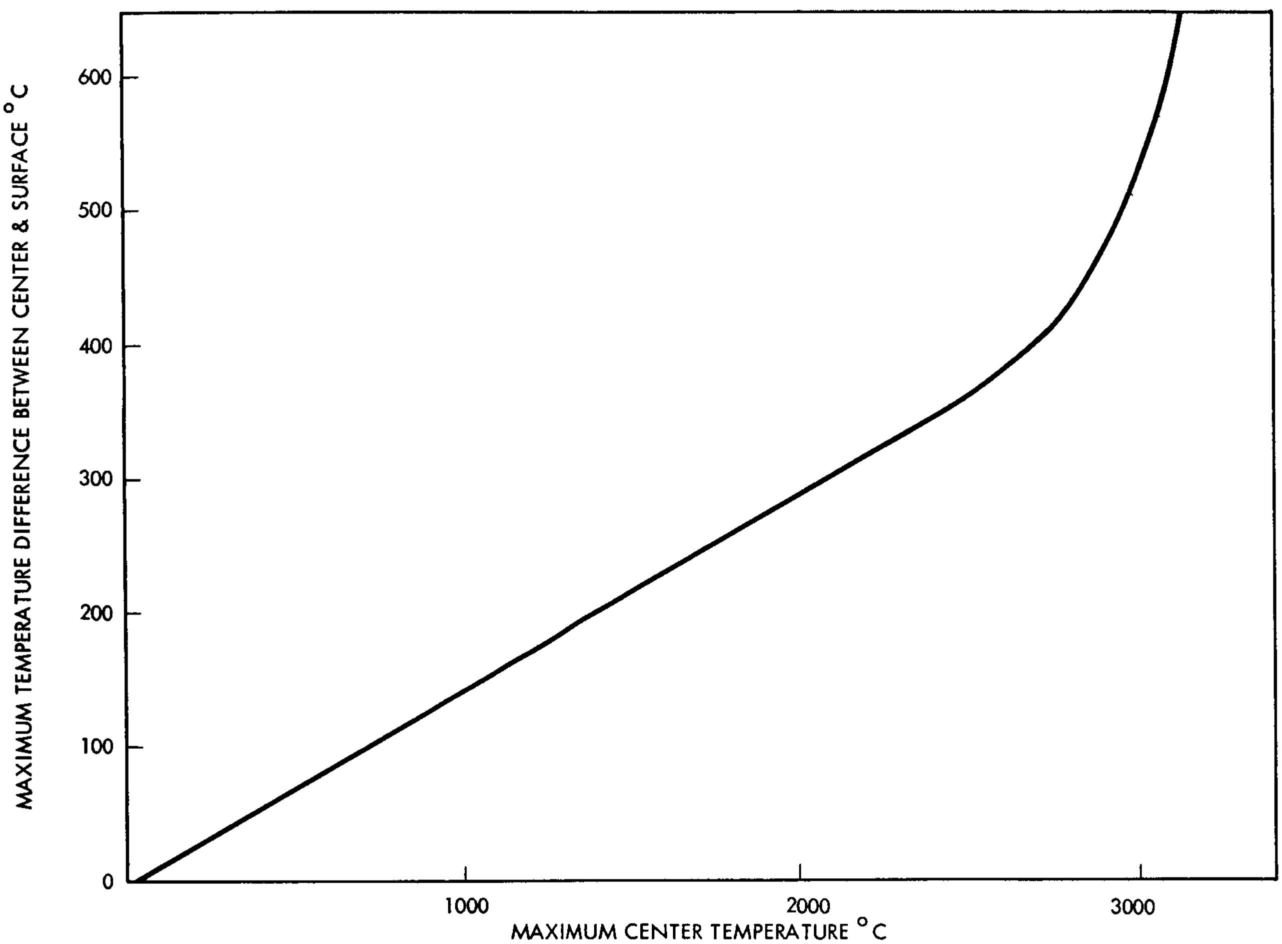

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FIGURE 4 - MAXIMUM TEMPERATURE GRADIENT IN NERVA FUEL MATERIAL

AS A FUNCTION OF MAXIMUM TEMPERATURE OF A TREAT PULSE cooling versus initial center temperature (maximum temperature difference) calculated by the method outlined above.

More detailed computer calculations were carried out to predict temperature distributions of NERVA fuel material during TREAT transients. An IBM 7094 computer was programmed for the "TOSS"** code to predict temperature as a function of time and radial position in 6-12 inch long hexagonal 19-hole NERVA fuel samples. The mathematical model consisted of dividing a unique segment of the hexagonal cross-section (1/12 part) into 232 nodes and assigning appropriate boundary conditions to nodes, node surfaces, and connectors. The physical model used was based on the following assumptions:

(1) Heat loss is by radiation from the fuel faces to a concentric graphite crucible inside a stainless steel autoclave.

(2) The fuel is infinitely long; i.e., no heat loss from the ends.

(3) The heat capacity and thermal conductivity of the fuel material is independent of temperature; mean values for C_p of 0.358 cal/(gm)($^{\circ}$C) and k of 0.250 w/(cm)(sec) were used.

(4) The graphite and UC_2 particles are homogeneously mixed such that there is uniform volumetric heat generation. The fuel material contains 19 w/o U.

(5) Uniform neutron fission density throughout the fuel volume. (An average value of 2.64×10^{12} fissions per gm of U-235 per mw-sec of TREAT power was used).

(6) Peak fuel temperature was limited to 3625°C to approximate the effect of graphite sublimation. Any input beyond this temperature was assumed to result in vaporizing graphite with a corresponding heat absorption of 170 k cal/mole.

(7) No heat absorption by UC_2 vaporization or by NbC liner melting.

Calculations were carried out using power versus time data from five typical TREAT transients. A summary of the computer results is given in Table 1. Figure 5

**David Bagwell, "TOSS an IBM 7090 Code for Computing Transient or Steady-state Temperature Distributions," K 1494, Union Carbide Nuclear Company

Table 1 - TREAT Transient Data Used In Computer Calculations

(Based on 19 w/o U In Graphite Irradiated In Standard Autoclave Capsules)

		Experimental Parameters				Calculated Values			
Computer Problem No.	TREAT Transient No.	Period (ms)	Integrated Power (Mw-sec)	Peak Power (Mw)	Heat Input To Fuel cal/gm	Max. Center Fuel Temp. °C	Center Fuel Temp. At Time Of Max. Temp. Diff.	Max. Fuel Temp. Diff. °C	Amount Graphite Vaporization, %
1	361	76	230	1060	745	2300	2240	330	
2	364	54	379	2430	1230	3625	--	--	.49
3	395	53	536	2410	1730	3625	3510	1000	4.1
4	365	40	645	3850	2090	3625	3510	1000	8.5
5	417	40	800	4465	2590	3625	3510	1000	13.1

shows a plot of TREAT reactor power versus time for the transients outlined in Table 1. Figure 6 presents a plot of center and apex temperature versus time for computer Problems 1 and 5. It is interesting to note that fuel material heat-up is essentially adiabatic to 1800°C. The plateau portion of Problem 5 curve is indicative of the extent of graphite vaporization. Figure 7 shows a plot of fuel temperature difference between fuel element center and surface points as a function of time for Problem 1. Figure 8 shows the same parameters for Problem 5 as a function of time after start of cooldown from 3625°C. These curves are an index of the thermal stress history during TREAT pulses. Figure 9 is a radial profile map of temperature distribution through fuel material at the time of maximum temperature gradient for Problem 5. Figure 10 shows a plot of this map along a radius through the apex and along a radius through the center of the fuel face.

Analysis of computer data reveals a number of interesting points. The steepest temperature gradient during radiation cooling is located on the fuel surface at the point of an apex. This corresponds to the coolest part of the surface. The amount of vaporization during an 800 Mw-sec transient has been calculated to be approximately 13.1%. This is in good agreement with TREAT experiments which have shown weight losses at this energy level of approximately 15%. Thus, it may be concluded that TREAT pulses in excess of those required to reach 3625°C will not increase thermal gradients; and thus, thermal stress levels in the fuel material.

Calculated temperature differences are approximately a factor of two higher than measured temperature differences. This can be partially explained by the time lag of thermocouples, by appreciable heat losses from the fuel sample ends and by the neutron flux depression in the fuel. The theoretical model did not account for any of these.

C. Results of Thermal Stress Experiments

Thermal stress experiments were carried out on NERVA reference fuel material employing three sample configurations. The first consisted of 1/2 inch diameter, 7-hole unlined, 3/4 inch long cylinders in standard autoclaves. The cylinders

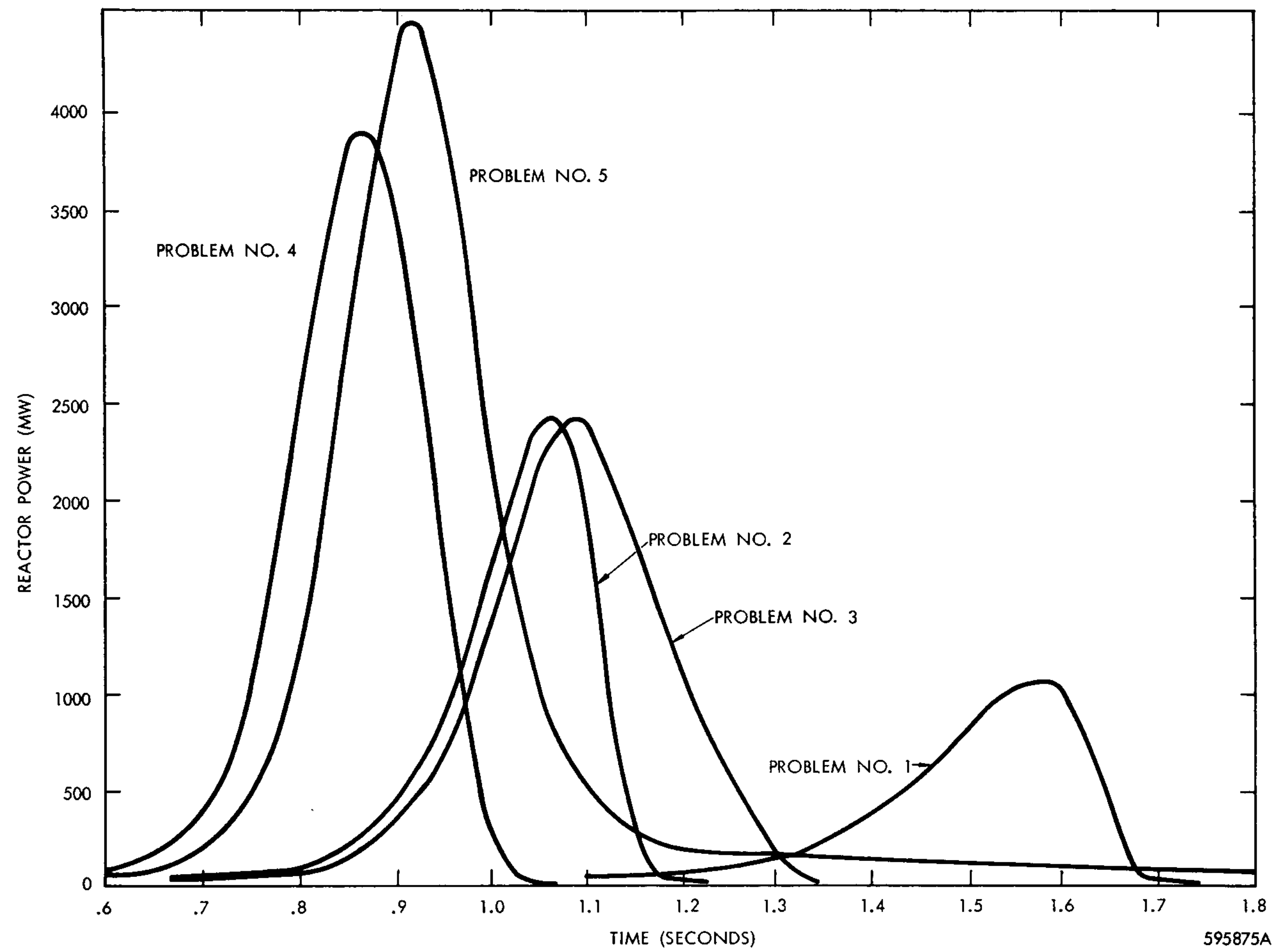

FIGURE 5 - TREAT REACTOR POWER VS TIME PROFILES

USED IN COMPUTER CALCULATIONS

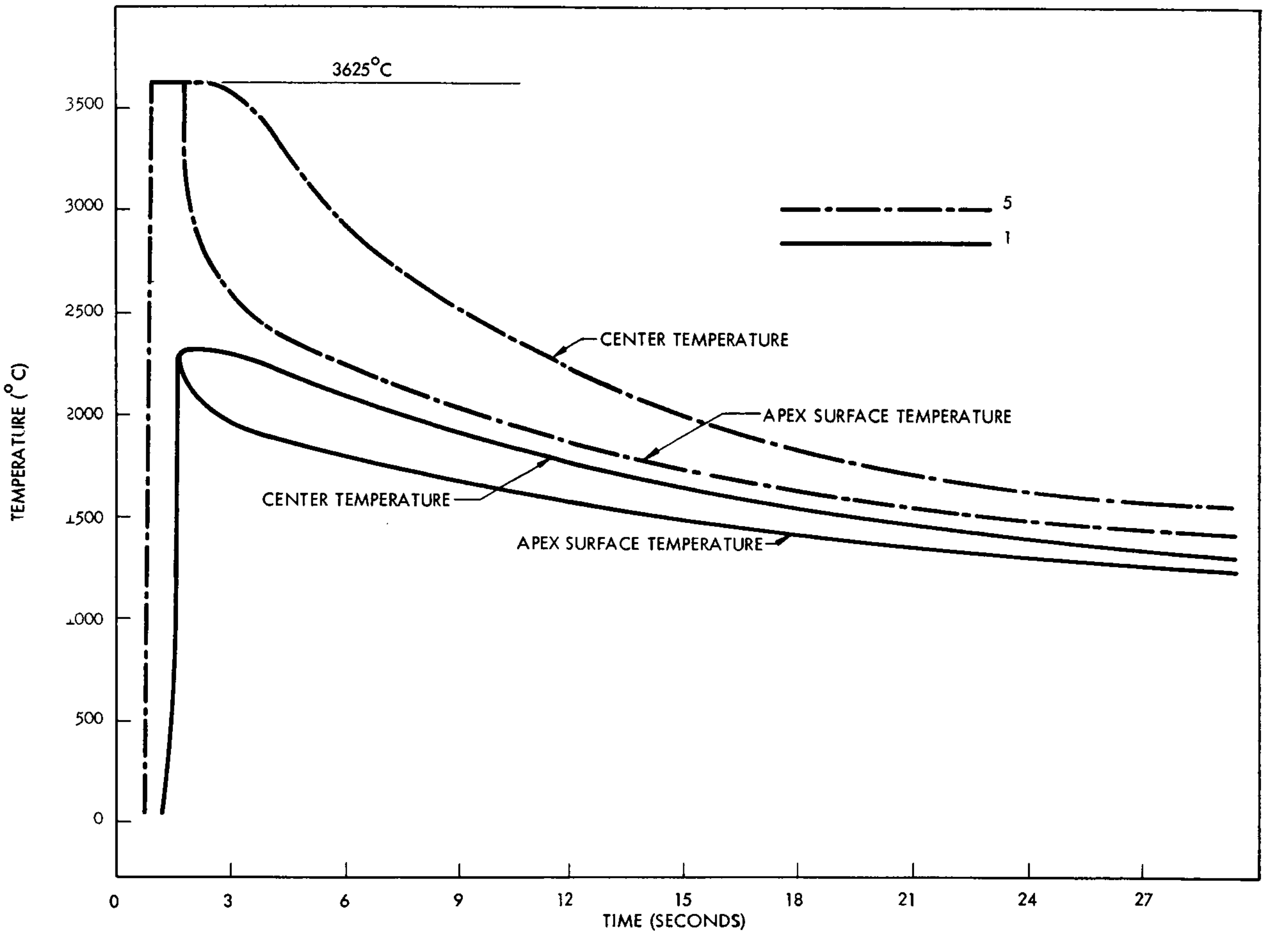

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FIGURE 6 - PREDICTED CENTER AND APEX SURFACE TEMPERATURE OF NERVA FUEL DURING TREAT TRANSIENTS

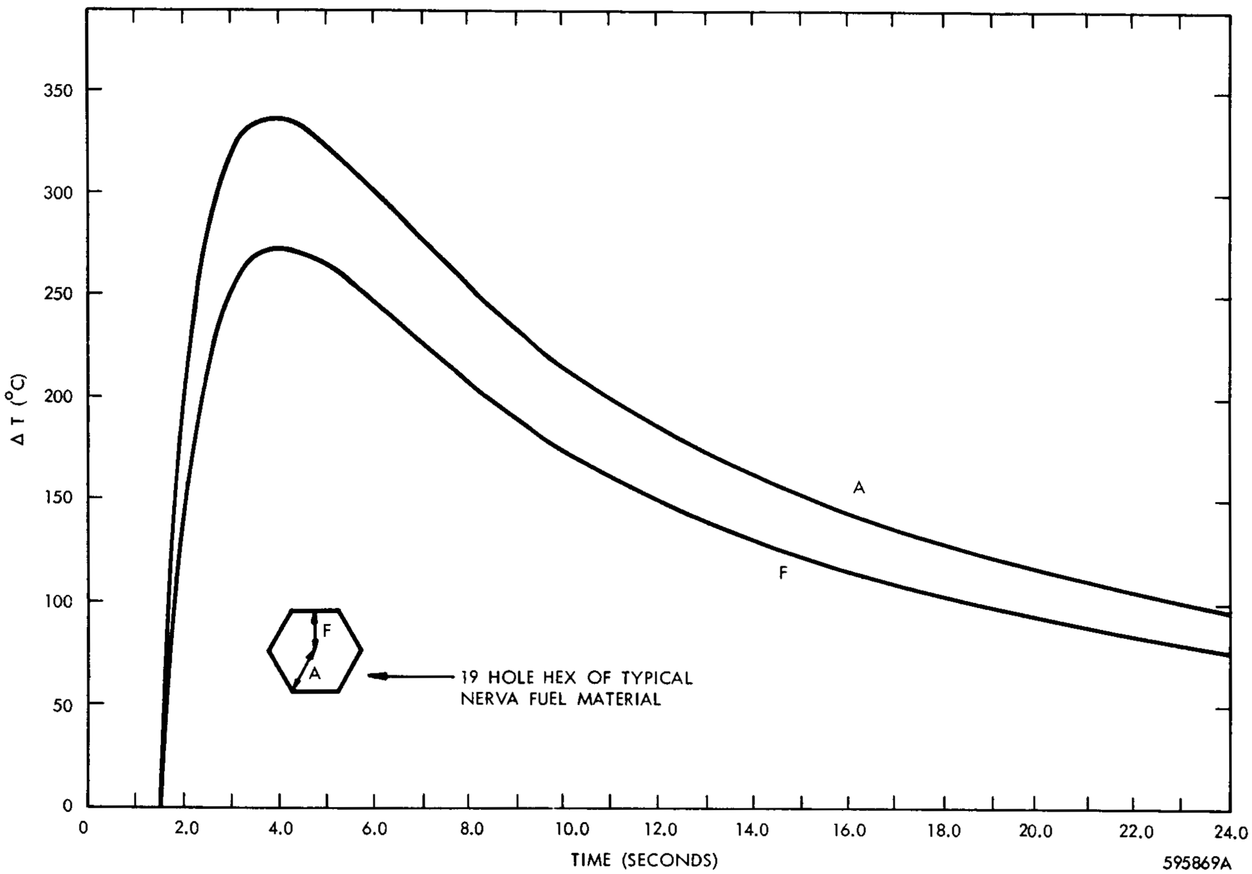

FIGURE 7 - TEMPERATURE DIFFERENCE BETWEEN CENTER OF HEXAGON AND SURFACE POINTS VS TIME AFTER BEGINNING OF TRANSIENT (PROBLEM 1)

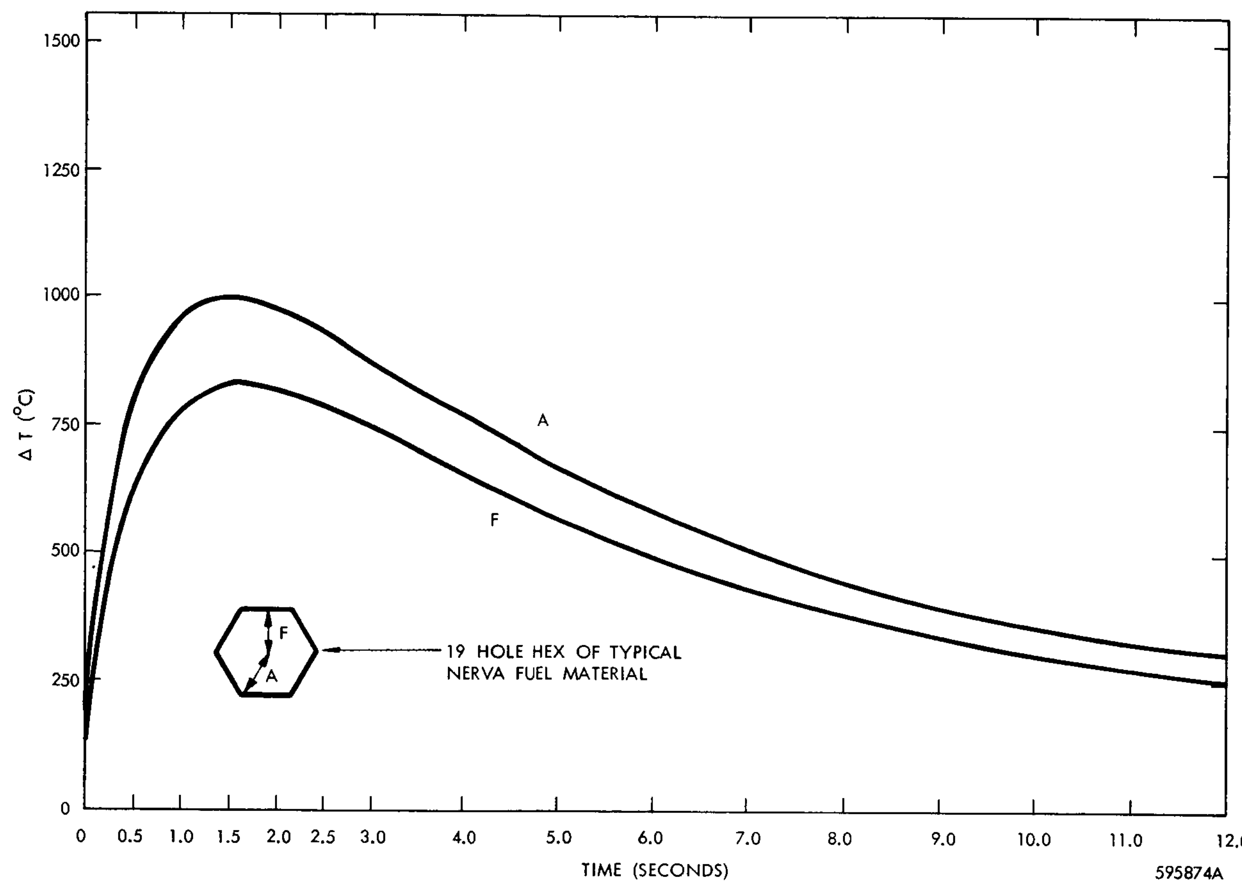

FIGURE 8 - TEMPERATURE DIFFERENCE BETWEEN CENTER OF HEXAGON AND SURFACE POINTS

VS TIME AFTER COOLDOWN FROM 3625°C (PROBLEM 5)

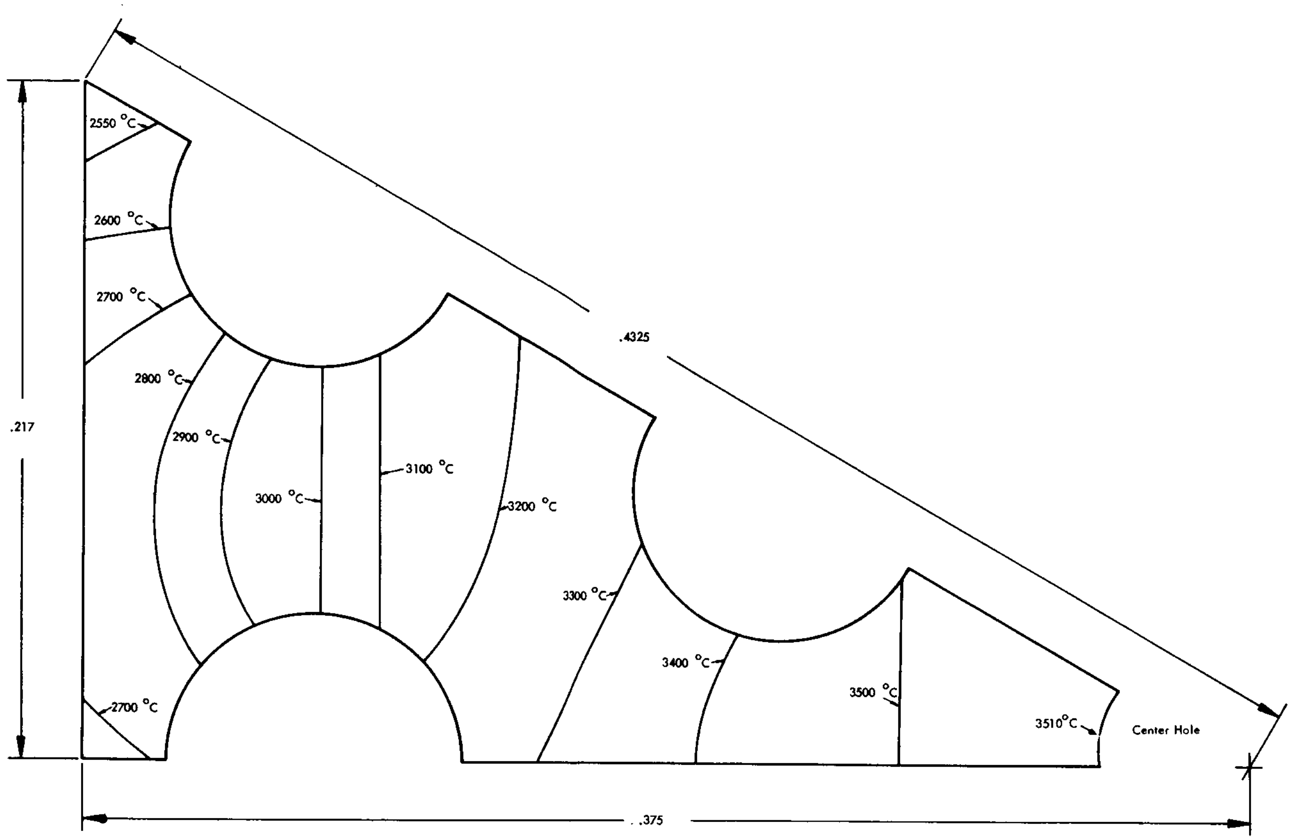

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FIGURE 9 - TEMPERATURE PROFILE INSIDE TYPICAL FUEL SECTION AT THE TIME OF MAXIMUM TEMPERATURE GRADIENT DURING COOLDOWN FROM 3625°C

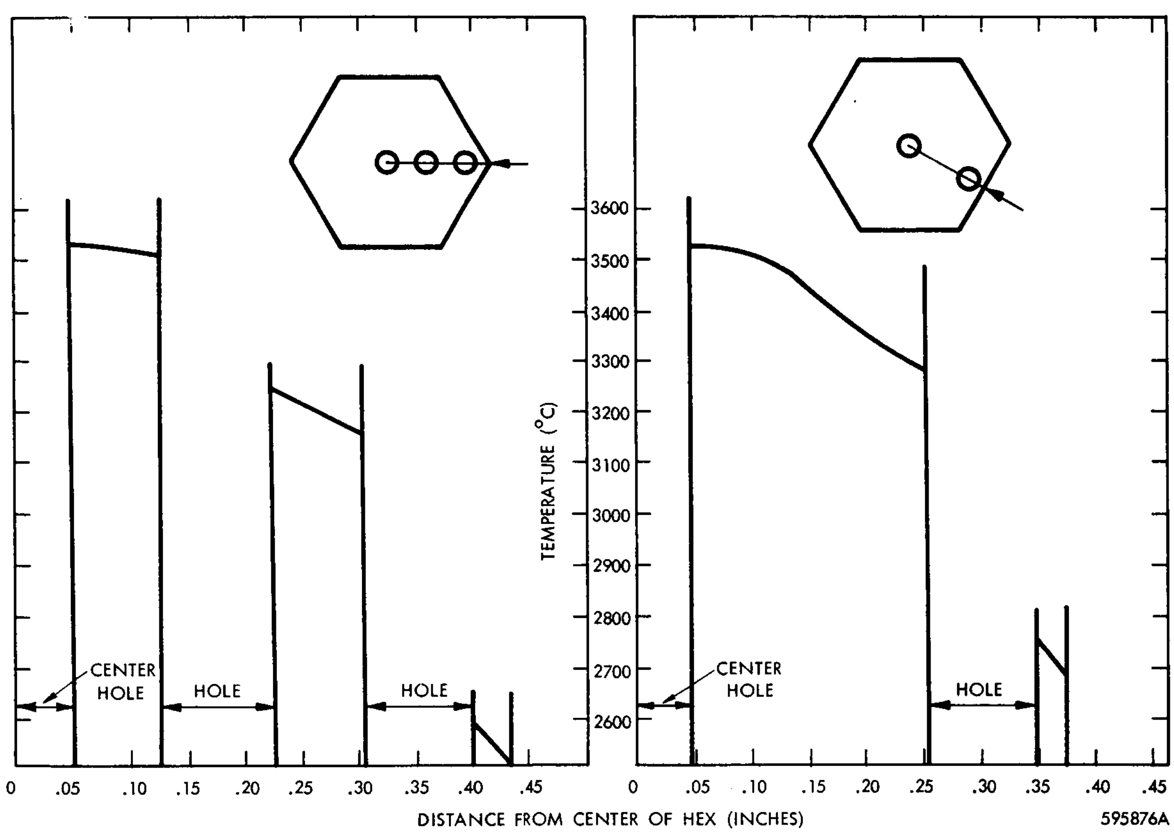


FIGURE 10 - TEMPERATURE DISTRIBUTION ALONG RADIUS THROUGH APEX AND CENTER FACE AT THE TIME OF MAXIMUM TEMPERATURE GRADIENT DURING COOLDOWN FROM 3625°C

were machined out of 19-hole hexagonal fuel elements. The second consisted of a 3/4-inch hexagonal, 19-hole NbC lined, 1-1/2-inch long sample in an autoclave capsule. The third consisted of 3/4-inch hexagonal 19-hole NbC lined, 3/4-inch long samples irradiated in photographic capsules.

Table 2 lists the irradiation conditions during these experiments. Figure 11 shows a plot of fuel sample temperature versus energy input in cal/gm of fueled graphite for various sample configurations.

D. Discussion of Thermal Stress Results

Table 2 indicates the cylindrical specimens exhibit thermal stress cracking (as shown on Figure 12) at energy inputs between 1300-1900 cal/gm. Figure 12 shows the increase in cracking with increasing temperature and temperature difference up to 2275°C and 120°C, respectively. CEN-129 which was pulsed to 2450°C and a temperature difference of 180°C resulted in less damage. Thus, maximum cracking occurred at an intermediate temperature. This phenomena may be related to the relief of stresses by plastic flow as the graphite temperature is increased. The hexagonal specimens (as shown on Figure 13) remained intact up to energy inputs of 2520 cal/gm. Figure 13 shows the behavior of hexagonal fuel samples irradiated under conditions which did not produce cracking (T-18 at 3100°C and ΔT of 610°C) and under conditions which did result in cracking (T-17 at 3350°C and ΔT of 700°C). It is interesting to note that the radial crack was located near an apex hole which the computer analysis predicted to be the region of steepest temperature gradient. (See Figures 9 and 10). This represents anomalous behavior since for the same energy input the hexagonal samples with larger radial dimensions develop larger temperature gradients than the cylindrical specimens.

The explanation for this difference in behavior may involve the internal stress configuration within the fuel material. There is evidence that the as-manufactured hexagonal fuel material contains residual internal stresses which result from radial variations of preferred orientation. X-ray studies indicate a high degree of preferred orientation (basal planes parallel to the axis of extrusion) at the surface of the fuel

Table 2 - Irradiation Conditions and Post-irradiation Observations of Thermal Stress Experiments in TREAT

TREAT Transient No.	Sample* Config-uration	Energy Mw-sec	Period ms	Energy Input cal/gm	Peak Temp.** °C	Maximum Center to Surface Temp. Difference	Post-irradiation Observations
CEN-126	1	430	59	980	1700 (E) 2650 (A)	90 (C)	Intact no visible cracks.
CEN-127	1	580	50	1320	2025 (E) 3350 (A)	105 (C)	Several large longitudinal cracks in webs
CEN-128	1	720	42	1660	2275 (E) 3625 (A)	120 (C)	Fractured longitudinally into three segments
CEN-129	1	820	40	1860	2450 (E) 3625 (A)	130 (C)	One small longitudinal crack on surface
CEN-148	2	490	57	1130	2380 (M) 3000 (A)	180 (M)	No visible cracks
T-15	3	300	76	1370	2360 (M) 3400 (A)	190 (M)	No visible cracks
T-16	3	388	70	1750	2600 (E) 3625 (A)	345 (C)	No visible cracks
T-18	3	548	52	2520	3100 (E) 3625 (A)	610 (C)	No visible cracks
T-17	3	644	48	2960	3350 (E) 3625 (A)	700 (C)	Radial crack on surface in webbing near apex of the hexagon

* Sample configurations:
1 - 1/2" diameter, 3/4 inch long fuel in standard autoclave
2 - 3/4" hexagon, 1-1/2 inch long fuel in standard autoclave
3 - 3/4" hexagon, 1-1/2 inch long in photographic capsule

** (A) Adiabatic sample temperature (maximum temperature of 3625°C).
(E) Estimated from calibration curves of measured temperature versus energy input (cal/gm).
(M) Measured with W-Re thermocouple and optical pyrometer.
(C) Estimated from calculations. Cylindrical samples were adjusted in proportion to the square of the diameter to put on the same basis as the hexagonal samples.

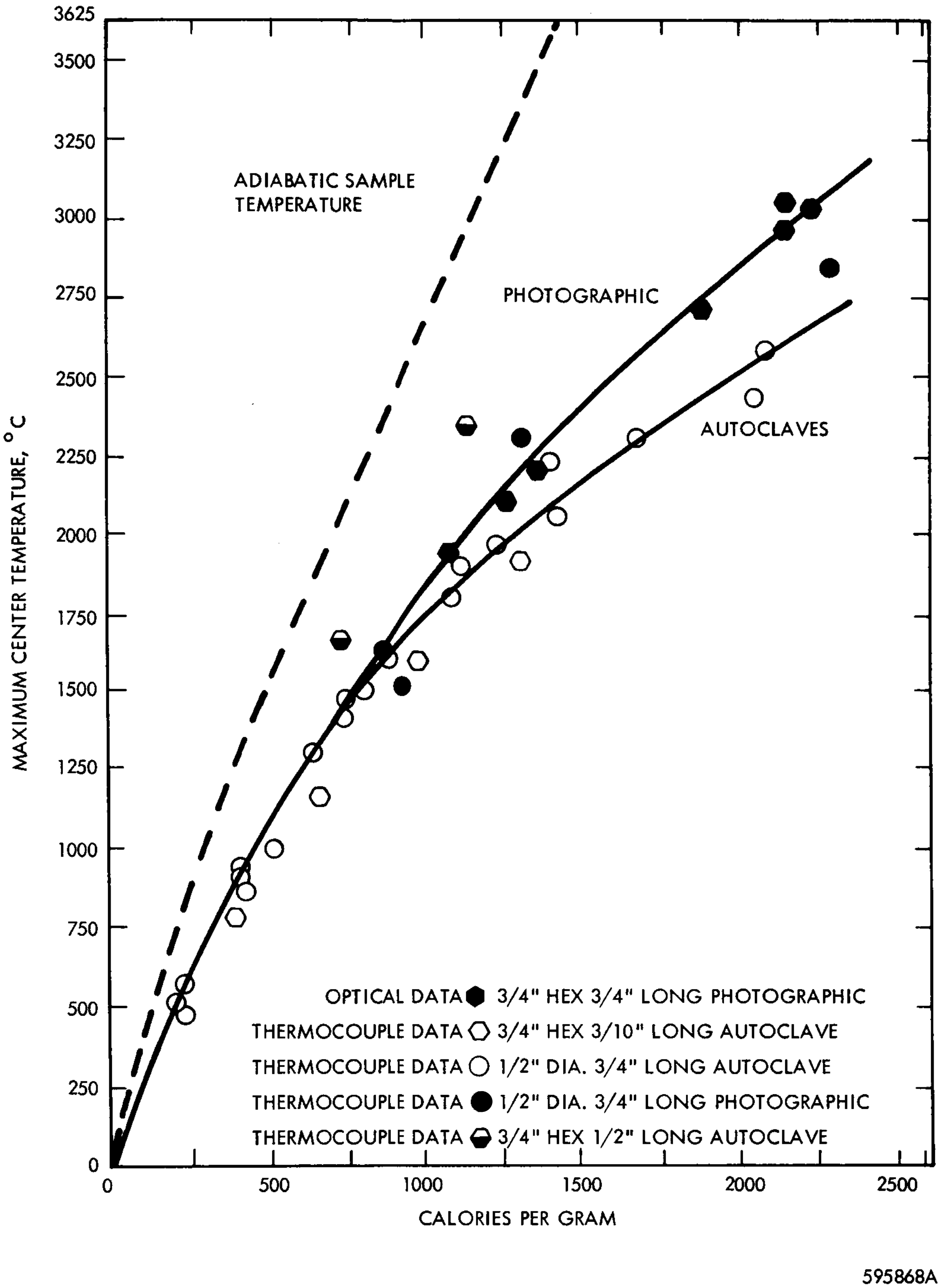


FIGURE 11 - FUEL SAMPLE TEMPERATURE VS ENERGY INPUT FOR VARIOUS SAMPLE CONFIGURATIONS (400 mg/cm^3 U LOADING)

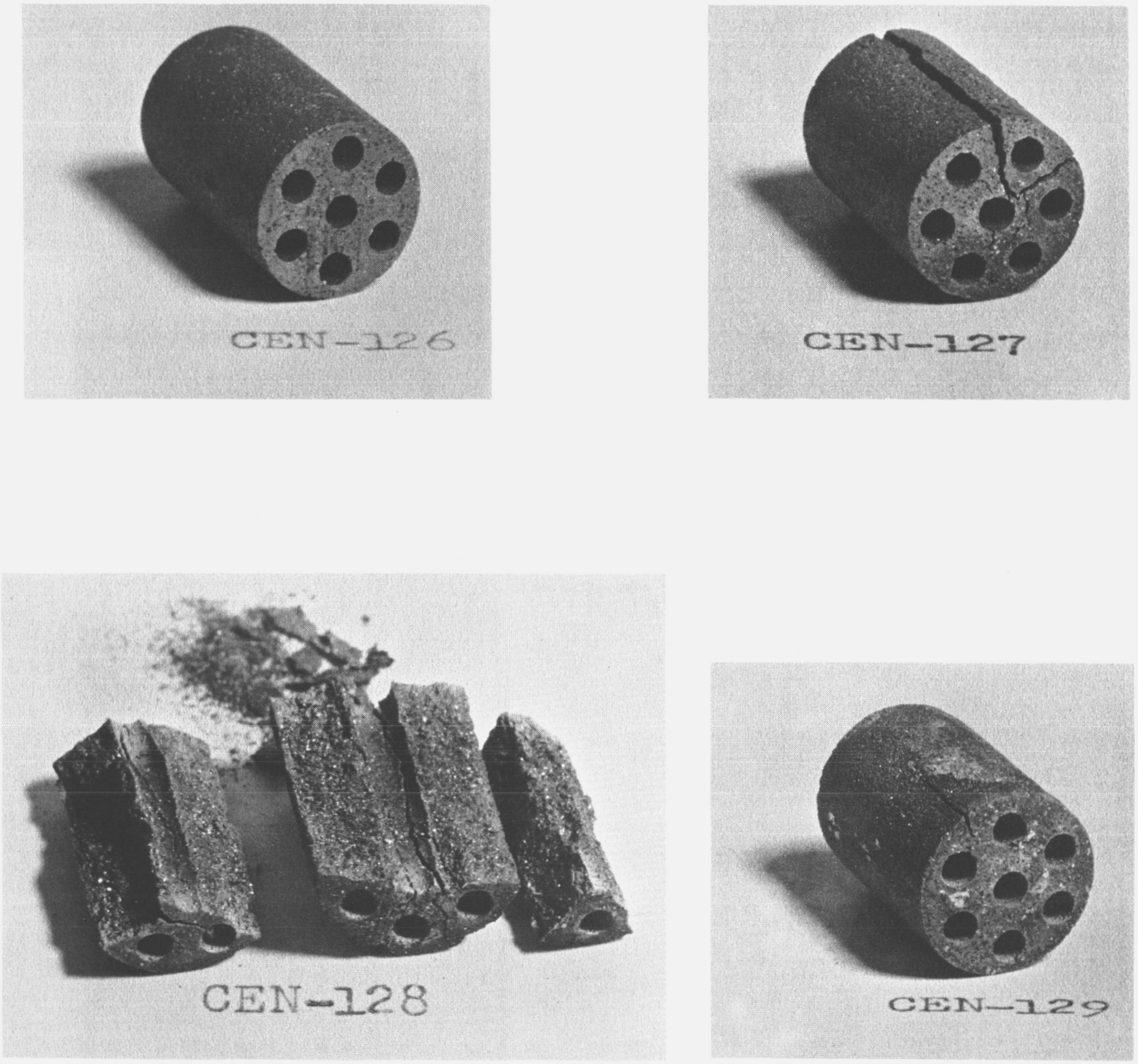
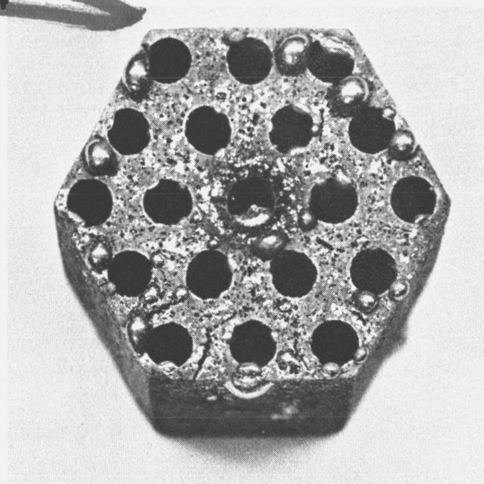


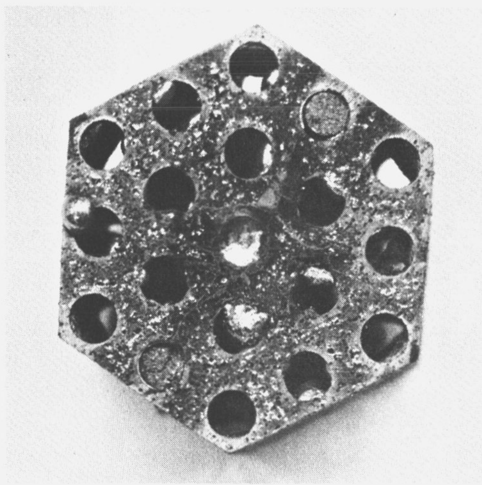
FIGURE 12 - CYLINDRICAL FUEL SAMPLES SHOWING THERMAL STRESS CRACKS AFTER IRRADIATION IN TREAT

CEN	Energy Input cal/gm	Max. Temp. °C	Max. ΔT °C
126	980	1700	90
127	1320	2025	105
128	1660	2275	120
129	1860	2450	130



T-17

Energy Input - 2960 cal/gm
Max. Temp. - 3350°C
Max. ΔT - ≈700°C
NbC Liner melted - No cracking



T-18

Energy Input - 2520 cal/gm
Max. Temp. - 3100°C
Max. ΔT - 610°C
NbC Liner melted - No cracking

FIGURE 13 - HEXAGONAL FUEL SAMPLES SHOWING THERMAL STRESS CRACKS AFTER IRRADIATION IN TREAT

element. The radial variation of preferred orientation gives rise to residual compressive stresses at the surface due to differences in coefficient of thermal expansion with preferred orientation. Thus, a hexagonal cross-section sample would appear to be stronger than a sample cut from the interior because of the residual compressive stress. The residual compressive stress on the hexagonal faces must be overcome by thermal stresses introduced by radiation cooling in order to produce a net resultant tensile stress. The cylindrical fuel samples were machined from the center of the hexagonal fuel material and would thus have little or no residual surface stress. Mechanical property studies on fuel elements are consistent with the proposed explanation.

E. Future Work

The work which has been completed indicates that an improved in-pile temperature history is desirable. In addition, it would be desirable to minimize the effect of heat losses from the fueled hexagon ends. To accomplish this, a series of experiments will be run on 6-inch long fuel samples. These samples will be instrumented with 12 tungsten-rhenium thermocouples located in selected radial and axial positions both in fuel holes and in the webbing between holes. Temperatures will be printed out on a high speed recorder as a function of time during transients. Pre- and post-irradiation examination will serve to define material behavior in terms of thermal gradients. Following the thermal stress experiments in a static helium environment, the work will be extended to an investigation of the effect of helium coolant through the fuel holes. This work will more closely mock-up the fuel stress pattern during NERVA operation.

F. Conclusions

Unconstrained NERVA fuel material (19-hole, hexagonal cross-section) is resistant to thermal stresses resulting from energy inputs up to 2520 cal/gm. This energy input gives rise to a maximum radial temperature gradient of at least 610°C between fuel center and surface during the cooling cycle. The material is remarkably resistant to thermal shock, having been heated from 25° to 3100°C in 0.5 seconds at maximum heating rates of 28,000°C/sec without damage to the graphite matrix. Microstructural changes occurred in coated particles and the NbC liner under conditions which did not affect the graphite matrix.

Experiments on 7-hole sections of the fuel material reveal anamolous behavior which appears to be related to the internal stress distribution within the fuel elements.

III. Steady-state Irradiation Experiments (GETR)

A. Experimental Techniques

Steady-state irradiations were carried out in the small (2 1/2 inch) trail-cable facility of the General Electric Testing Reactor. The trail-cable facility provides access to the reactor pool by means of a 30 foot long tube. Capsule experiments attached to long lead tubes may be inserted, adjusted, and removed during reactor operation. Figure 14 shows the pertinent features of this facility. Figure 15 shows the design of the initial capsules used for NERVA fuel material irradiations. These capsules permitted irradiation of twelve 1/4 inch diameter, 1/4 inch high cylinders at temperatures up to 2200°C. Temperature was monitored with a tungsten-rhenium thermocouple and with temperature monitor wires. Flux was measured with Co-Al flux monitor wires.

The experimental procedure involved insertion of the capsule to a pre-selected axial position in the trail-cable facility which was chosen to give the desired fuel temperature. A temperature recorder monitored the experiments and permitted adjustment of capsule position to higher or lower flux positions as required. After temperature equilibrium was reached, the irradiations were carried out for fifteen minutes at a flux of 1×10^{14} n/(cm^2)(sec).

B. Experimental Results

Two NERVA fuel material irradiations (FP-3 and FP-4) were carried out in the General Electric Testing Reactor. These experiments were designed primarily to evaluate the release of fission products from NERVA fuel as a function of pretreatment, irradiation temperature, and post-irradiation heat treatment. The discussion which follows will be concerned only with the fuel material behavior with respect to uranium migration from UC_2 particles as a result of irradiation.

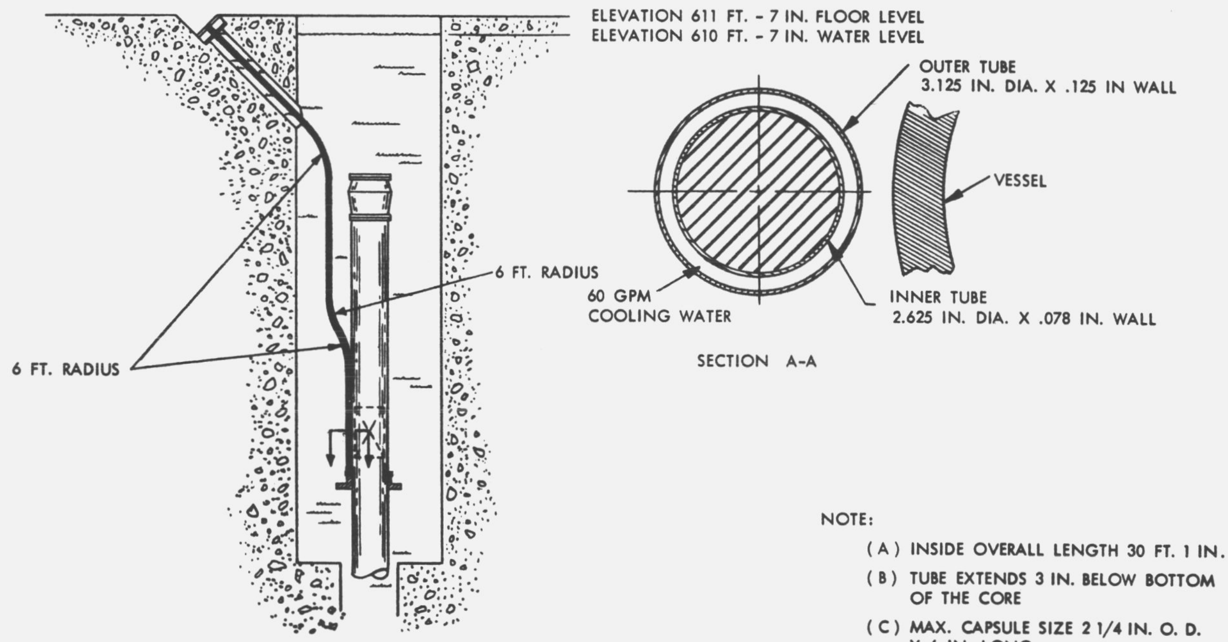


FIGURE 14 - GETR TRAIL-CABLE FACILITY

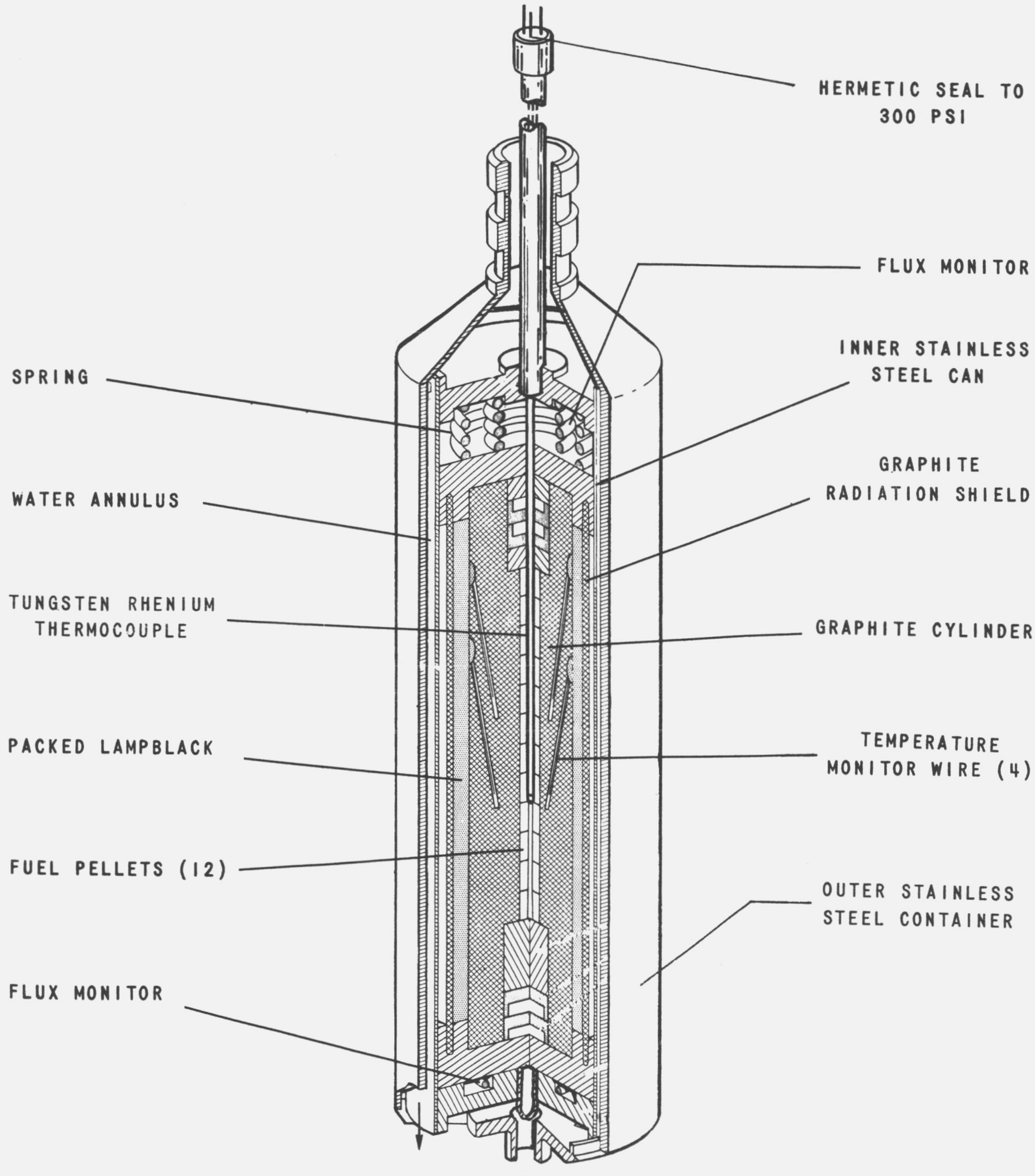


FIGURE 15 - FP-3 AND FP-4 IRRADIATION CAPSULE

Each capsule contained twelve 1/4 inch diameter by 1/4 inch high hollow cylindrical fuel pellets with an inside diameter of 0.095 inch*. The pellets contained 0.4 grams of U-235 per cubic centimeter of fueled graphite as UC_2. The fuel particles were 100-150 microns diameter UC_2 particles coated with 25 microns of pyrocarbon coating. All fuel pellets were leached in nitric acid to remove UC_2 exposed during the machining of the pellet surfaces. The pellets were then subjected to two different heat treatments. Type 1 was degassed for several hours at 1900°C; whereas, Type 2 was heated for fifteen minutes at 2600°C in a helium atmosphere to affect significant migration of uranium from the coated fuel particles. Irradiation parameters for the two capsules are shown in Table 3.

Preliminary metallographic examination of the irradiated pellets suggests that some migration occurred as a result of irradiation of the Type 1 material at 1800°C for fifteen minutes. In addition, there is some indication of additional migration of the previously heat treated Type 2 material. Migration of uranium in Type 1 material was not expected at as low a temperature as 1800°C. Additional metallographic studies of the irradiated material are in progress.

C. Future Work

Figure 16 shows the new T-T series capsules which are now in process of being assembled. These will permit irradiation of three 1/2 inch diameter, 1 inch long cylinders of fuel material. They are instrumented with up to nine tungsten-rhenium type thermocouples to monitor temperature. Thermal insulation is provided by a graphite sleeve and by gas gaps. Heat is dissipated in the same manner as in the previous capsule design, that is by forced convection from the metal capsule surface to the GETR process water. The neutron flux will be monitored with Co-Al flux wires.

Six capsules are now in process of being assembled for subsequent irradiation at the General Electric Testing Reactor. The purpose of these irradiations will be to

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* The fuel pellets were machined from 19-hole hexagonal fuel material.

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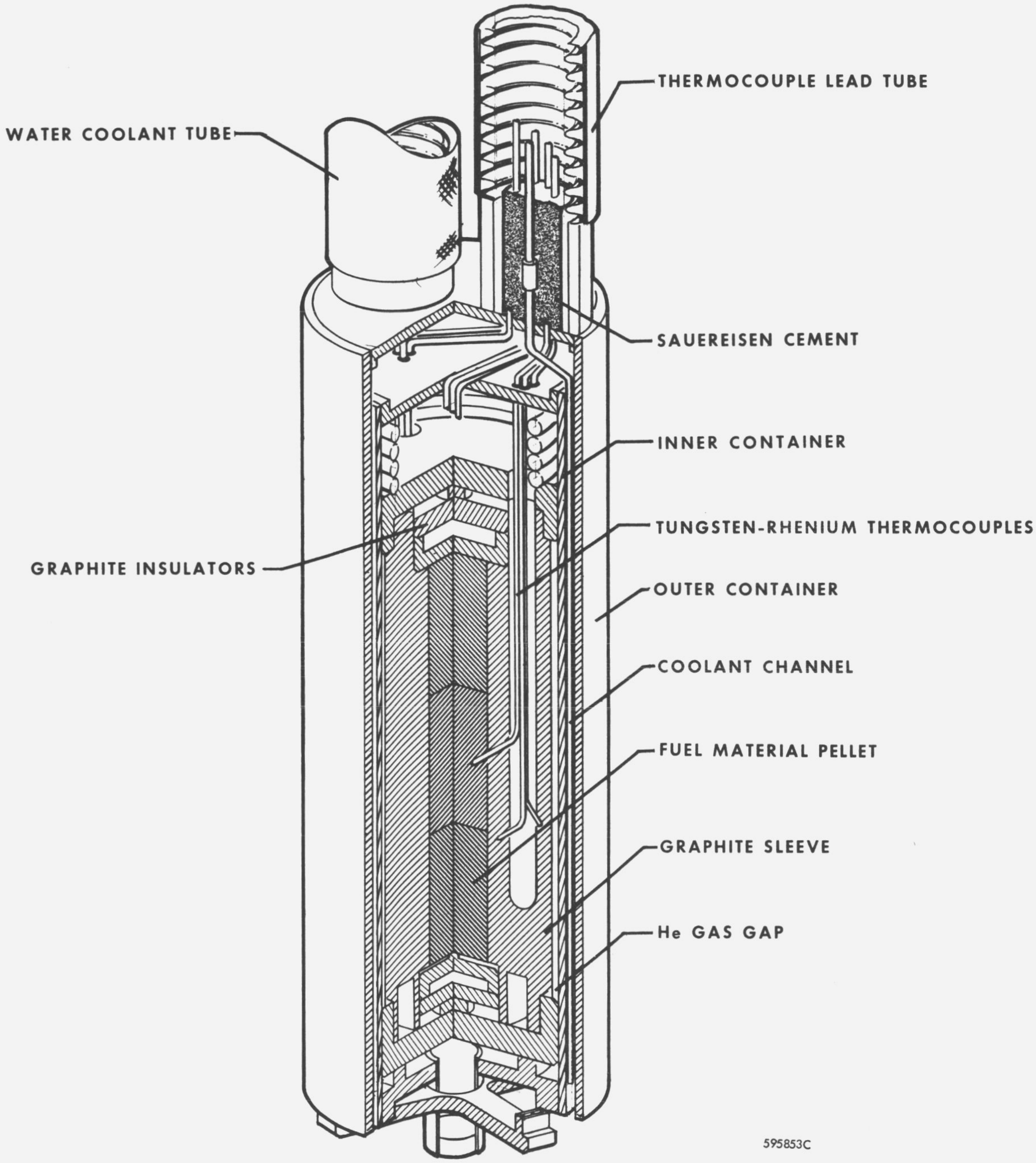

FIGURE 16 - TT - SERIES IRRADIATION CAPSULE

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Table 3 - Irradiation Parameters for FP-3 and FP-4

		Irradiation Conditions		Neutron Flux
Capsule No.	Fuel Type	Measured Temp., °C	Time Minimum	$n/(cm^2)(sec)$
FP-3	6 pellets Type 1	1800	15	1.1×10^{14}
	1 disc Type 1			
	6 pellets Type 2			
FP-4	12 pellets Type 1	2175	15	1.2×10^{14}

study the behavior of NERVA fuel material as a function of irradiation temperature and time and as a result of thermal and chemical pretreatment. The temperature range which will be investigated will be 2100°-2500°C and the irradiation times will correspond to the burnup levels achieved in 5-40 minutes of NERVA reactor operation. The fuel material to be studied includes outgassed reference material, fuel exposed to flowing hydrogen at 2400°C for five minutes, and material partially hydrolyzed by heating at 2400°C for 1/2 hour and holding at 100°C in 100% relative humidity atmosphere for one day.

D. Conclusions

The preliminary studies indicate that irradiation enhanced uranium migration may occur in 15 minutes at 1800°C. However, further work is required before definitive conclusions may be drawn relative to fuel particle behavior.

Acknowledgements

The TREAT experiments were carried out by Messrs. W. Yario and C. R. Simmons while on assignment from Westinghouse Astronuclear at Argonne National Laboratory. The operation of the TREAT reactor was performed by Messrs. J. Bolandand H. Lawroski of the Idaho Division of Argonne National Laboratory. The IBM computer work was carried out by Mr. R. Patterson at WANL.